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Title: European characterization factors for damage to natural vegetation by ozone in life cycle impact assessment

Authors: Thomas M. W. J. van Goethem^{1*}, Philipp Preiss², Ligia. B. Azevedo¹, Joachim Roos², Rainer Friedrich², Mark. A. J. Huijbregts¹, Rosalie van Zelm¹

¹ Department of Environmental Science, Institute for Water and Wetland Research, Radboud University Nijmegen, P.O. Box 9010, 6500 GL, Nijmegen, the Netherlands

² Department for Technology Assessment and Environment (TFU), Institute for Energy Economics and the Rational Use of Energy (IER), Universität Stuttgart, Hessbruehlstr. 49a, 70565 Stuttgart, Germany

*Corresponding author: e-mail: tgoethem@science.ru.nl. Tel: +31-243652060 and fax: +31243553450

19 **Highlights**

20 * Characterization Factors expressing damage to natural vegetation by ozone were derived

21 * Emissions and deposition of NO_x and NMVOC in 65 European regions were included

22 * Characterization factors were largest for NO_x emissions in Southern European regions

23 * NO_x contributes for 81% to ozone damage in natural vegetation in Europe

24 * NO_x contributes more to ozone than to acidification effects on natural vegetation

25

ABSTRACT

Spatially explicit characterization factors (CFs) for tropospheric ozone damage on natural vegetation caused by anthropogenic NO_x and NMVOC emissions are presented for 65 European regions. The CFs were defined as the area-integrated increase in the potentially affected fraction (PAF) of trees and grassland species due to a change in emission of NO_x and NMVOCs. The CF consists of a Fate Factor, quantifying the relationship between the emission of precursor substances and ozone exposure, and an area-integrated Effect Factor, quantifying the relationship between ozone exposure and the damage to natural vegetation. The relationships describing the ecological effects of a pollutant were based on a lognormal relationship between the PAF and ground level ozone concentration. We found higher CFs for NO_x compared to NMVOC, and these were largest in south European regions. Furthermore, we found that both the fate factor and effect factor contribute to the spatial differences found in the CFs. Our study shows that effects caused by ozone exposure from NO_x emissions are larger than those of acidification caused by NO_x, indicating the importance of including ozone effects to natural vegetation in life cycle assessment studies.

Key words: *Atmospheric Fate Factor; NO_x and NMVOC; AOT40; trees and grassland; Effect Factor; Species Sensitivity Distribution*

1. INTRODUCTION

Long term surveys show that tropospheric ozone background concentrations have significantly increased over recent decades, and concentrations are predicted to further increase with 0.5 – 2% per year over the next 50 years in the Northern Hemisphere (Vingarzan et al. 2004, Derwent et al 2007). Tropospheric ozone in a given area can have several sources, such as downward transport of stratospheric ozone to the troposphere or by photochemical reactions of nitrogen oxides (NO_x) and non-methane volatile organic compounds (NMVOCs). NO_x and NMVOC are primary precursor substances originating from anthropogenic and non-anthropogenic emissions. These pollutants can come from local sources or long-range transport (Ainsworth et al. 2012). Ozone is recognized as an important air pollutant, affecting human health and vegetation, including trees and grassland species (Ashmore 2005). Adverse effects in plants include reduction of growth and seed production, premature senescence, reduced ability to withstand stressors, and increased leaf injuries (Emberson et al. 2003).

In life cycle impact assessment (LCIA), characterization factors (CFs) estimate the environmental impact of a pollutant per unit of emission (Udo de Haes et al. 2002). Although CFs are available for human health damage caused by ozone (e.g. Van Zelm et al. 2008), studies assessing the impact to natural ecosystems have yet only included regionalized fate and exposure modeling, excluding effects on natural vegetation (Bare., 2011, Hauschild et al. 2006, Frechette-Marleau et al. 2008). Recently, Van Goethem et al. (2013) developed quantitative exposure-effect relationships for ozone on natural vegetation (forests and natural grasslands, respectively). These relationships can be used to include ozone effects on natural ecosystems in LCIA.

The aim of this study was to determine region-specific characterization factors for damage on natural vegetation of tropospheric ozone caused by anthropogenic NO_x and NMVOC emissions. The CFs were characterized for 65 European regions and subsequently compared to assess the differences in impact between the regions. Furthermore, normalization factors for ozone exposure on natural vegetation were presented. The normalization factor equals the potentially affected fraction of natural plant species in Europe due to emissions of NO_x and NMVOC in 2010 per capita.

2. METHODS

2.1 Characterization factors

The characterisation factors were defined as the area-integrated change in Potentially Affected Fraction (PAF) of forest and natural grassland species due to a change in emission of ozone precursor substances, i.e. NO_x or NMVOC (in m².yr/kg). The CF consists of a Fate Factor (FF), quantifying the relationship between the emission of precursor substances and ozone exposure, and an Effect Factor (EF), quantifying the relationship between ozone exposure and the damage to natural vegetation. Ozone exposure is expressed as the sum of the differences between the hourly mean ozone concentration and 40 ppb during daylight hours over the relevant growing season (AOT40 in ppm.h). The CFs for ozone were calculated for 65 European regions separately as;

$$CF_{x,i,e} = \sum_j \sum_e (FF_{x,i \rightarrow j} \cdot EF_{j,e}) \quad (1)$$

where FF_{x,i→j} (ppm.h.yr/kg) is the partial fate factor representing the change in AOT40 in receiving grid j (spatial resolution of 0.5 x 0.5 degrees) following a change in the emission of substance x (i.e. NO_x and NMVOCs) in region i and the effect factor EF_{j,e} (m²/ppm.h) is the change in the PAF of species of vegetation e (i.e. trees and grasslands) in grid j due to a change in ozone exposure.

2.2 Fate factor

The partial fate factor ($FF_{i \rightarrow j}$, unit: ppm.h.yr/kg) represents the change in AOT40 in a receiving compartment cell j ($\Delta AOT40_{i,j}$, unit: ppb.h) due to a change of emission of precursor x in region i (ΔM_i , [kg/yr]):

$$FF_{x,i \rightarrow j} = \frac{\Delta AOT40_j}{\Delta M_i} \quad (2)$$

The exposure is taken over time and for daytime only (Tuovinen, 2000). The AOT40 exposure index is a measure of chronic ozone exposure widely used in the risk assessment of ozone (LRTAP, 2004).

Partial fate factors for the European continent were determined with the EMEP atmospheric chemical transport model, which simulates emissions, atmospheric transport, chemical transformation, and removal from air of NO_x and NMVOCs and estimates ground level ozone concentrations (Tarrasón, 2009a). To calculate FFs for the grassland vegetation, the change in AOT40 on 1 m ground level height was used. For the trees vegetation the upper canopy height (3 m) was used. The model divides Europe into 65 emission source regions (EMEP, 2008), and receptor grid cells of 0.5°x0.5°. To derive the partial fate factors, emissions of NO_x and NMVOCs are decreased by 15% compared to the baseline emission inventory for each region. The 15% represents a realistic “quasi-marginal” change of emissions but still allows to assume sufficient linearity and to downscale the change of impacts to a unit of emission change (Tarrasón 2009b). FFs were determined for each region, precursor pollutant, and 2010 background emissions. The emission data set for 2010 corresponds to the baseline Current Legislation (CLE) scenario, developed by IIASA for the development of the Thematic Strategy on Air (Amann et al., 2008; Tarrasón, 2009b). Because of inter-annual variability in the meteorology, average results based on

meteorological years 1996, 1997, 1998, and 2000 were derived as these years represent typical conditions (Tarrasón, 2009a).

2.3 Effect factor

EFs were derived via the following steps. First, species-specific AOT40 exposure-biomass response functions, as reported by Van Goethem et al. (2013), were used to derive EC50 values for trees and grassland species. The species-specific EC50 equals the AOT40 at which there is a 50% reduction in biomass compared to a situation with no ozone over-exposure, i.e. AOT40 = 0. We selected the EC50, as it follows the same approach employed for toxicity in LCA (see e.g. Rosenbaum et al., 2008). Note that some species showed to be insensitive to ozone exposure, i.e. no EC50 value was derived for these species. In a second step, we used the EC50-values to derive a Species Sensitivity Distribution (SSD) for respectively forest and natural grassland species, taking into account the fraction of species with no biomass decrease. An SSDs represents a cumulative stressor-response distribution based on single-species sensitivity data. Assuming a lognormal species sensitivity distribution for ozone exposure, the PAF can be derived as:

$$PAF_{j,e} = \frac{1 - f_{nbd}}{\sigma_e \cdot \sqrt{2 \cdot \pi} \cdot AOT40_{j,e} \cdot \ln 10} \cdot \int_0^{AOT40} \exp\left(-\frac{1}{2} \cdot \left(\frac{\log(AOT40_{j,e}) - \mu_e}{\sigma_e}\right)^2\right) dAOT40 \quad (3)$$

where AOT40_{j,e} represents the ambient ozone concentration in grid j of vegetation type e (either forest or natural grassland), μ_e is the average of the ¹⁰logEC50 values for ozone in AOT40-units (ppm.h), as observed for different species in vegetation type e, f_{nbd} is the fraction of species with no biomass decrease and σ_e is the standard deviation of the ¹⁰logEC50-data within vegetation type e.

In a third step, we calculated the marginal change in PAF due to the marginal change in ground level ozone exposure (in ppm.h), equal to the derivative of equation 3, via:

$$\frac{\partial PAF_{j,e}}{\partial AOT40_j} = \frac{1 - f_{nbd}}{\sigma_e \cdot \sqrt{2 \cdot \pi} \cdot AOT40_{j,e} \cdot \ln 10} \cdot \exp \left(-\frac{1}{2} \cdot \left(\frac{\log(AOT40_{j,e}) - \mu_e}{\sigma_e} \right)^2 \right) \quad (4)$$

In a final step, the grid-specific marginal effect factor (MEF) per vegetation type was defined as:

$$MEF_{j,e} = \frac{\partial PAF_{j,e}}{\partial AOT40_j} \cdot A_{j,e} \quad (5)$$

where $A_{j,e}$ is the area (m^2) occupied by vegetation type e in grid j.

The AOT40_{j,e} data were based on grid-specific background AOT40 concentrations for 2010 determined by the EMEP model (Simpson et al., 2003). For the grassland vegetation, AOT40 values based on a growing season of May-July and a ground level height of 1m were used. For the trees vegetation, April-September and the upper canopy height (3 m) was used. The Global Land Cover 2000 (GLC2000) database was used to calculate the grid-specific area occupied by each vegetation type (Bartholomé and Belward, 2005). Classification of GLC2000 types into trees and grasslands can be found in table S1.

2.4 Sensitivity analysis

To test the sensitivity of the CF regarding the relationship describing the ecological effects of ozone on natural vegetation, the marginal effect factor (MEF) was compared to two other options to calculate an effect factor. This was done because there is no consensus yet in the best way to derive an EF (Huijbregts et al., 2011).

A simplified EF, assuming a linear change in PAF with changing AOT40 represents the average effect between a PAF of 0.5 and 0. This linear method is commonly used in ecotoxicology (Pennington et al. 2004) :

$$LEF_{j,e} = \frac{\Delta PAF_{j,e}}{\Delta AOT40_j} \cdot A_{j,e} = \frac{0.5 \cdot (1 - f_{nbd})}{10^\mu} \cdot A_{j,e} \quad (6)$$

where 10^μ is the AOT40 value that affects 50% of the species in vegetation type e in grid j.

For the average effect factor (AEF), it is assumed that the distance between the current and desired situation is proportionally distributed over the stressor range. And, as opposed to LEF, the actual concentration of AOT40 in grid j ($AOT40_j$) estimates the average distance to the PAF at 0 (Huijbregts et al., 2011). The average distance between the current state and the preferred state of the environment can be calculated as:

$$AEF_{j,e} = \frac{\Delta PAF_{j,e}}{\Delta AOT40_j} \cdot A_{j,e} = \frac{PAF_{j,e}}{AOT40_j} \cdot A_{j,e} \quad (7)$$

where $PAF_{j,e}$ is related to the $AOT40_{j,e}$ representing the ambient ozone concentration in grid j of vegetation type e.

2.5 Normalization factor

The normalization factors (NF) for ozone impacts in Europe were derived by multiplication of the region-specific characterization factors with the substance-specific emissions in each region of 2010, divided by the total population in all regions (Table S2) (Verstreng et al., 2012). The normalization factor equals the area-integrated potentially affected fraction of natural plant species in Europe due to emissions in 2010 of NO_x and NMVOC per capita (in PAF.m²/capita):

$$NF = \frac{\sum_x \sum_i (M_{x,i} \cdot CF_{x,i})}{\sum_i (N_{pop,i})} \quad (8)$$

where NF is the normalization factor of the summation of all 65 regions. $M_{x,i}$ is the emission of precursor x (NMVOC or NO_x) in region i (in kg/yr), $CF_{x,i}$ is the characterization factor for substance x in region i and $N_{pop,i}$ is the number of inhabitants in region i (CIESIN, 2005).

2.6 Statistical analysis

Pearson correlation test was used to determine linear correlation between CF and total Fate Factor to have an indication if the FF contributes most to the variation in CF results for emissions of both NMVOC and NO_x.

3. RESULTS

3.1 Species Sensitivity Distributions

Figure 1 shows the species sensitivity distributions for grassland species and trees based on EC₅₀-data with the fraction of species with no biomass decrease included. The SSDs were based on 87 grassland species and 9 tree species. The percentage of species in the dataset that exhibited a biomass reduction was 71% for grassland species and 100% for tree species.

Figure 1

3.2 Characterization factors

Characterization factors for damage to natural vegetation by tropospheric ozone were calculated for 65 European regions both for NMVOC and NO_x (in m².yr/kg) (Fig. 2). The full region-specific results are given in the SI (Table S3). The region-specific CFs and emission weighted CFs for Europe are included in table S3. Weighing was done based on region specific emissions in 2010 (table S2) (Verstreng et al., 2012). The CFs for ozone damage due to NMVOC emissions range from 0.3 to 5.0 m².yr/kg, with smallest CFs for Sweden and Finland and largest for Luxembourg and Italy. The CFs for NO_x emissions range from -0.3 to 20.6 m².yr/kg, with smallest a negative CF for the Netherlands and largest CFs for France and Switzerland. The negative CFs for NO_x indicate that increased emissions will actually lead to a net reduced ozone exposure. Overall, the CFs for NO_x emissions are largest in South European regions. For NMVOC emissions CFs are largest for regions in Central Europe.

Figure 2

The correlation between CF and the total FF was tested . This was done by plotting the characterization factors versus the sum of the partial fate factors (\sum FF) for every

emission region (Fig. 3). A correlation between CF and \sum FF was found by using the Pearson correlation test, r-values of 0.73 and 0.70 were found for NMVOC and NO_x respectively. Both r-values were reported with a p-value <0.0001. However, the R-squares of the linear regressions, 0.53 and 0.49 for NMVOC and NO_x, imply that both the fate and effect factors contribute significantly to the spatial differences found in the CF.

Figure 3

Variability in EF is determined by variability in (1) the grid-specific area covered by each vegetation type and (2) grid-specific AOT40 values (see fig. 4). The area covered by grassland and forest varies across Europe (fig. S1). Therefore, the relative contribution of the grassland and forest types to the EF varies by grid (fig. S2). Variability in FF is also determined by the grid-specific area covered by each vegetation type, because the change in ozone exposure is determined for a different height per vegetation type (fig. S1,S2).

3.3 Normalization factor

The normalization factor for ozone impact on natural vegetation due to emissions of NO_x and NMVOC in 2010 is $1.4 \cdot 10^{-10} \text{ m}^2 / \text{capita}$. NO_x contributed 80% and NMVOCs 20% to the normalization factor, implying that NO_x is the main contributor to damage by ozone exposure in natural vegetation in Europe.

3.4 Sensitivity analysis

The different EF types for both forests and natural grasslands are plotted against the grid-specific background AOT40 values in Figure 4, along with the frequency of AOT40 background exposure values in Europe. Most AOT40 exposure values range from 0.01 to 1

ppm.h. In this range the LEF is generally 2 orders of magnitude larger than both the MEF and AEF. Here, AEF is also larger than the MEF. In higher AOT40 ranges, 10 to 100 ppm.h, the MEF is larger than the LEF and AEF

Figure 4

The linear effect factor method results, on average, in CFs that are a factor of 3 larger than the CFs calculated with the marginal effect method. On the other hand, the average effect factor method results in CFs that are typically a factor of 7 smaller compared to CFs calculated with the marginal effect method. Correlation between the CFs based on the marginal approach versus the CFs based on the linear and average approach was large (Pearson correlation test, r^2 -values of 0.81 and 0.89, respectively. See Figure S3).

4. DISCUSSION

In this study, characterization factors representing damage to natural vegetation by ozone exposure were calculated for unit emissions of NMVOC and NO_x in 65 European regions. Furthermore, normalization factors were calculated, which showed the potentially affected fraction of plant species over a certain area due to European emissions of NMVOC and NO_x per capita. In the following, we discuss the benefits and limitations of the calculation procedure and provide an interpretation of the results obtained. Furthermore, our work is compared to another impact category, i.e. damage due to ozone is compared to damage due to acidification.

4.1 Uncertainties

4.1.1 Fate factor

The Unified Eulerian EMEP model has been selected to derive fate factors for ozone exposure. Validation studies of the EMEP model show that EMEP gives a description of

ozone formation in Europe that is in good agreement with the much more comprehensive IVL chemistry model and observational data (Andersson-Sköld and Simpson, 1999, Hov et al. 1978, Simpson 1992). Furthermore, intercomparison studies of atmospheric chemistry-transport models, comparing EMEP with, e.g. the LOTOS-EUROS and TM5 model, indicate that the EMEP model gives results, which are in line with measured summer daytime averages, maxima and the diurnal cycle, which are important for modeling AOT40(Vautard et al., 2007; Van Loon et al., 2007).

Uncertainty in the derivation of fate factors for ozone formation relates to the complex non-linear chemistry of photochemical ozone creation, including the interaction between the precursors NO_x and NMVOC, and meteorological conditions (Simpson et al., 2003; Solberg et al. 2004). Emission inventories are a large source of uncertainty in model predictions (Atkinson, 2000). Meteorologically induced variability of AOT40 shows a gradient decreasing from north-west to south-east Europe (EEA, 2009). The variability was estimated as approximately 10% for southern Europe, 20–30% in central Europe and 50% or more in the United Kingdom (EEA, 2009). Further uncertainty arises due to the fact that different NMVOC substances have different potential to create ozone (Andersson-Sköld and Holmberg 2000). The mixture of different NMVOCs can differ across regions and sectors. However, the current FF is an average for the common mix of NMVOCs. The model can be improved by including sector dependent NMVOC speciation or even substance-specific model runs (Derwent et al., 2007).

Because only the EMEP regions are covered, receptor areas outside this area are not taken into account in the fate factor calculations. As a result, the area-integrated ozone exposure will be underestimated for NMVOC and NO_x emissions, because impacts due to ozone outside of the receptor area are not taken into account.

4.1.2 Effect factor

The GLC2000 database was used to calculate the grid-specific area occupied by each vegetation type. However, not all GLC2000 land cover classes corresponded in terms of species composition to our classification in vegetation types, therefore making the right allocation for some of the classes uncertain, especially for transitional vegetation (Table S1). For instance, the shrub cover class was appointed to the grassland type but it might contain tree species as well. Making a more detailed division in vegetation types, however, was not possible because of lack of detailed response data for a wide range of taxonomic groups.

The relationships between ozone exposure and damage for forests and grasslands were calculated using species-specific exposure-response data based on experimental studies (Van Goethem et al., 2013). However, in contrast to Van Goethem et al. (2013) no distinction was made between annual and perennial grassland species, because no grid-specific data on the area each species group occupied was available. A general concern regarding the exposure-response data is that the sensitivity to ozone exposure can be overestimated at the community level due to a bias towards the use of sensitive species in fumigation experiments, therefore leading to overestimated CFs (Mills et al., 2007). Furthermore, there are uncertainties regarding variation in atmospheric conditions and species responses (Fuhrer, 2002). In literature significant inter- and intraspecific variation in response to ozone exposure was reported for species occurring in multiple regions (Oksanen et al., 2001). This variation is mostly explained by differences in climate, for instance, climatic factors such as high vapour pressure deficits can reduce ozone uptake through stomata (Biswas et al, 2008). This implies that region-specific SSDs are needed to show the distinctly different environmental conditions and species assemblages, e.g. on an ecoregion or biome level. This was, however, not possible due to lack of available data. (Van Goethem et al., 2013). Furthermore, there is larger uncertainty in the tree SSD because it was based on 9 tree species compared to 61

species for grassland. However, the EF for grassland has a higher contribution to the CF compared to the EF for forests, therefore limiting overall uncertainty (fig. S4). The receptor grid resolution and sizes also influence the EF. For example, the EF results get more accurate at higher resolution when the vegetation is heterogeneously distributed in the grids. Besides the AOT40 approach, which is based on ozone concentrations only, another index is currently in use for indicating risk of ozone damage to natural vegetation in Europe. This approach, i.e. the stomatal ozone flux or Phyto-toxic Ozone Dose (POD), involves estimating the amount of ozone entering via the stomata of vegetation, which, on biological grounds, would yield more elaborate response models (Mills et al., 2011). Comparison between these approaches indicates a different ozone exposure distribution across Europe (Simpson et al., 2007). Currently, however, there are not enough species-specific flux-based exposure-effect models available to derive EFs and subsequent CFs based on the POD approach.

The marginal approach to calculate effect factors was used as the default in our study, the interest in LCIA generally lies in assessing the influence of small emission changes (Van Zelm et al., 2009; Struijs et al., 2011). However, alternative approaches found in the literature are linear effect factors (LEFs) and average effect factors (AEFs) (Huijbregts et al., 2011). The linear approach is commonly used in other impact categories, such as toxicity, in case no reliable background pollutant concentrations are available (Rosenbaum et al., 2008). As the linear approach disregards the shape of the exposure-response curve, it is considered to be less reliable compared to the marginal or the average approach. The AEF represents the average distance between the current and the preferred state of the environment per unit of emission (Huijbregts et al., 2011). The virtue of the AEF approach is that it focuses on the total change of ozone exposure that is required to reach the preferred state of the environment. The correlations found between the CFs based on the different effect factor methods imply that they largely result in the same ranking between regions and substances

(fig. S3). Looking at the absolute CF values, the choice for either the marginal or average effect factor method gives a typical difference in the CFs of a factor 7. This implies that the choice for a marginal or an average approach has a substantial influence on the CFs. Therefore, when comparing to other impact categories, e.g. toxic impacts, for which the approach is different the choice for an effect factor model is important.

4.2 Interpretation of the results

Tropospheric ozone formation is driven by complex non-linear photochemistry between the precursor substances NMVOC, NO_x and background tropospheric ozone (Atkinson, 2000; Butkovic et al., 1990). For instance, at nighttime and in the immediate vicinity of very large emissions of NO, ozone concentrations are depressed through the process of NO_x titration (Sillman, 2003). This consists of the removal of ozone through reaction with NO. Generally, summer ozone levels in Europe show an increasing gradient from the north-west to the south-east part. In the Mediterranean, large scale circulation cells are established and coastal emissions can be trapped for several days in land-sea breeze circulation systems (Borrego et al., 1995). These effects in relation with larger solar radiation levels in southern Europe can result in high ozone levels. This is also reflected in our results, where characterization factors for NO_x are largest in south European regions, which are the result of larger AOT₄₀ levels per unit of emission (FF) and consequently also higher EFs. The CF for NO_x is on average 3.5 times larger than the CF for NMVOC. These findings correspond with the results of Hauschild et al. (2006), who found larger CFs for NO_x by a factor of 3.6. The negative characterization factor in the Netherlands for NO_x indicates that increased emissions will actually lead to reduced ozone formation. This is caused by the titration effect (Simpson et al., 2003). The normalization factor for ozone impact also indicates that NO_x is the main cause of ozone damage to natural vegetation. This difference is caused by both larger CFs

and larger emissions for NO_x, i.e. the emissions for NO_x are on average 1.5 times larger compared to those for NMVOC.

The ILCD handbook recommends to provide CFs for carbon monoxide and methane as well (EC-JRC, 2011). However, the importance of carbon monoxide and methane in ozone formation compared to NMVOC and NO_x is much lower (Wayne, 2000), hence the potential contribution of these substances to tropospheric ozone formation was not addressed in this paper.

Our results for NO_x can be compared with those for acidification by Van Zelm et al. (2007) and Roy et al. (2013). The CF for NO_x related to acidification of European forests of Van Zelm et al. (2007) was 0.37 m²·yr/kg for a time horizon of 500 years. Our emission weighted CF for ozone damage to forests by NO_x emissions in Europe was 3.4 m²·yr/kg. Roy et al. (2013) reported a European acidification CF for NO_x of 5.0 m²·yr/kg, while our CF for ozone damage of NO_x is 5.6 m²·yr/kg. These results indicate a larger effect due to ozone damage by NO_x emissions than that of acidification by NO_x emissions, showing the importance of including ozone damage to natural vegetation in LCIA. This result is confirmed in literature as the contribution of NO_x emissions to ground-level ozone formation, acidification, and eutrophication is considered of comparable importance (Johansson et al., 2001). Furthermore, ozone is also identified as an important pollutant with regards to ecosystem damage when compared to acidification (Ashmore, 2005).

Summarizing, we determined spatially explicit characterization factors for damage of tropospheric ozone by anthropogenic NO_x and NMVOC emissions on natural vegetation in 65 European regions. It is the first time that CFs for ozone were derived for impacts on natural vegetation on an endpoint level. Fate factors were derived with the EMEP atmospheric chemical transport model. The effect factors were based on a lognormal relationship between the potentially affected fraction of species and ground level

375 AOT40. We found that NO_x is the major contributor to damage caused by ozone.
376 Furthermore, we found that both the fate and effect factors contribute to the spatial
377 differences found in the CFs. Our study shows that effects caused by ozone exposure
378 from NO_x emissions is larger than that of acidification caused by NO_x.

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385 **SUPPLEMENTARY INFORMATION**

386 **Table S1:** Categories GLC2000 database

387 **Table S2:** Emissions of NMVOC and NO_x per region in 2010

388 **Table S3:** Characterization factors European regions in 2010

389 **Figure S1:** Area of grassland and forest in each grid.

390 **Figure S2:** Comparison area grassland and forests

391 **Figure S3:** Comparison characterization factor based on different effect models

392 **Figure S4:** Comparison effect factors for grassland species and trees

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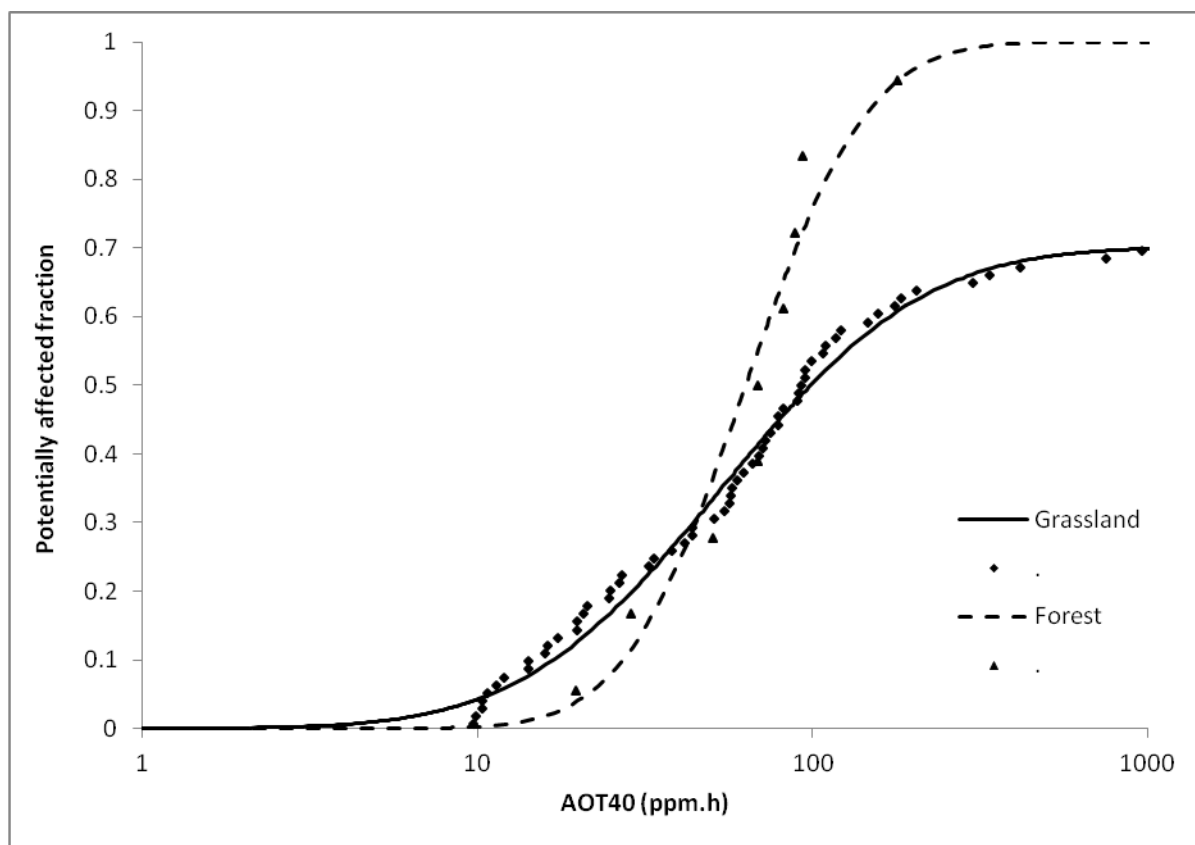
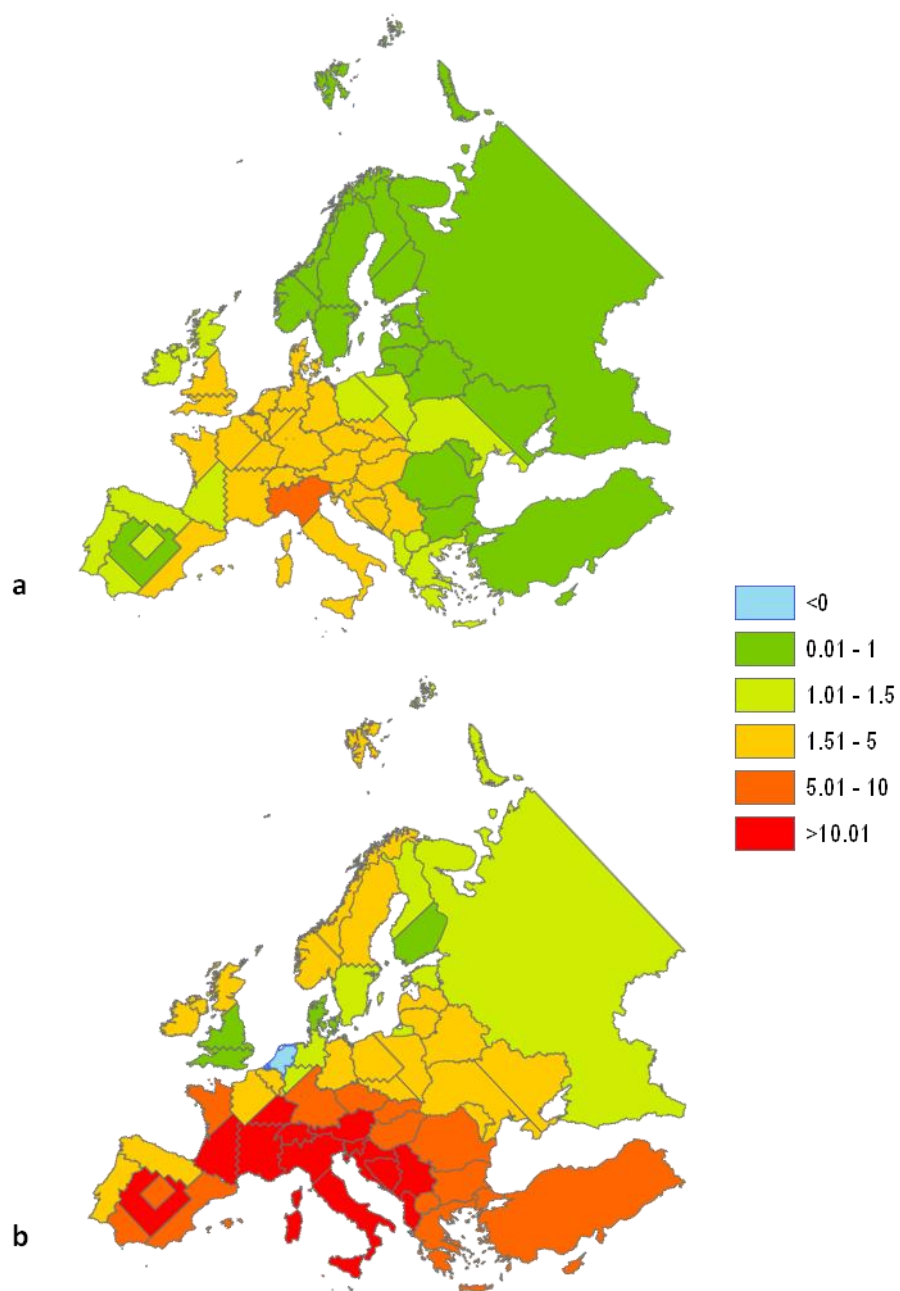


Figure 1. Species sensitivity distributions based on EC50 values for trees (n=9) and grassland (n=87) using a lognormal distribution.



539

540 **Figure 2. Region specific characterization factors for damage due to ozone caused by**
 541 **emissions of (a) NMVOC and (b) NOx (in m².yr/kg).**

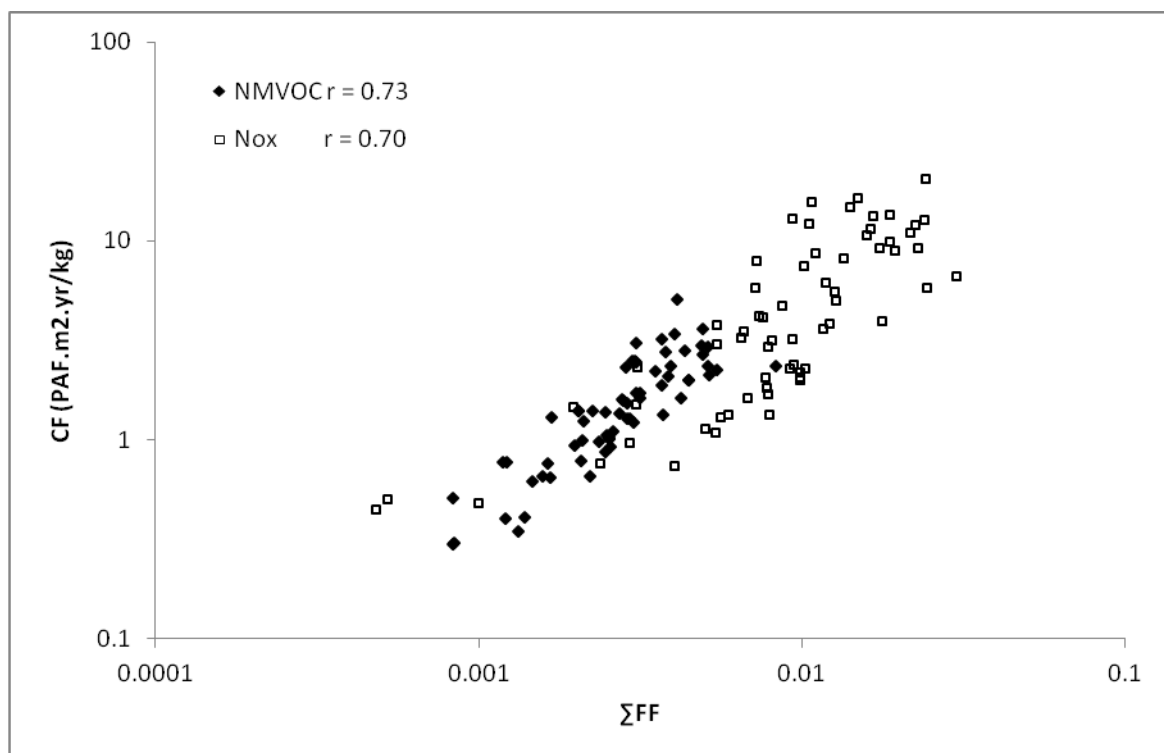
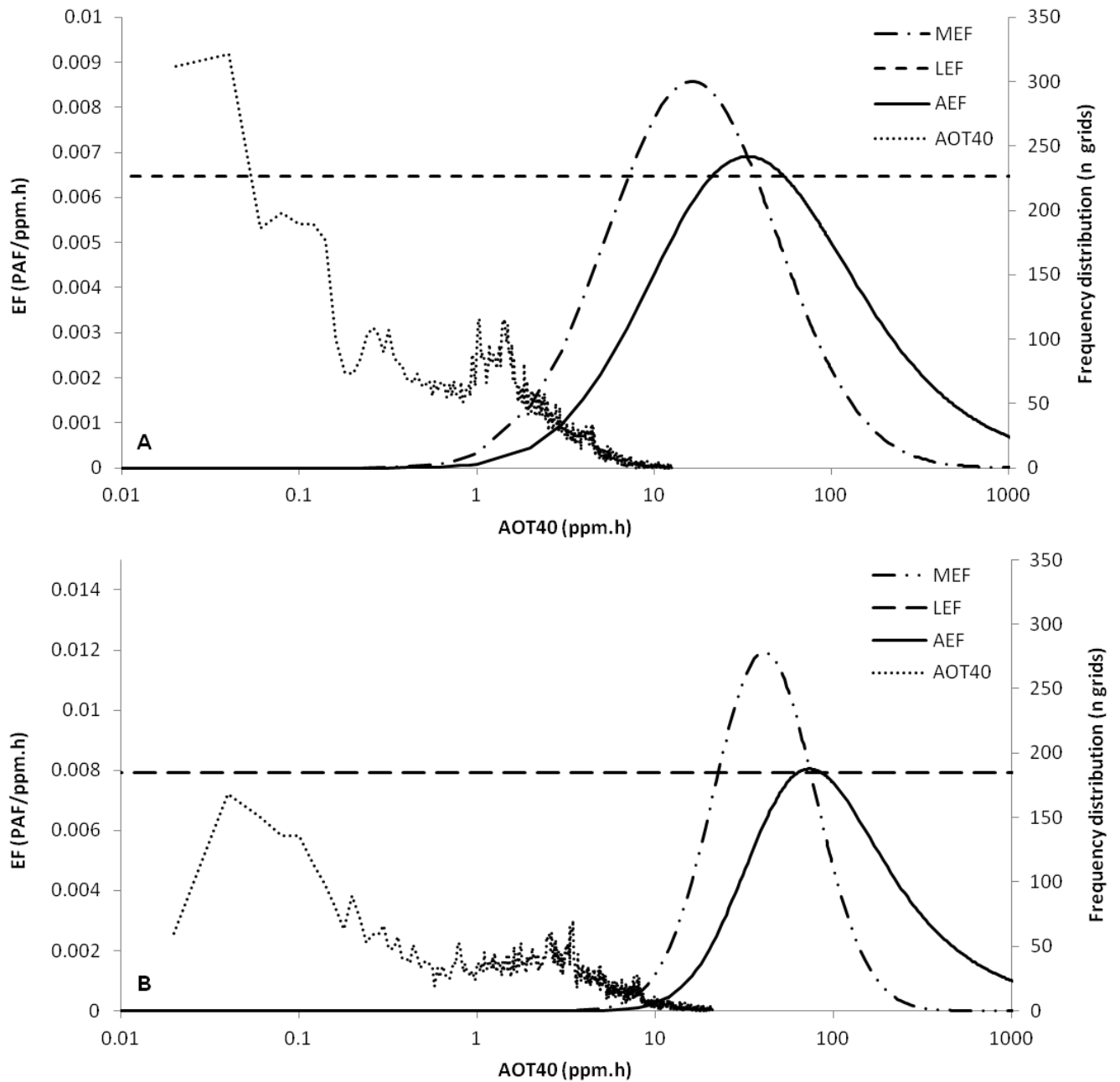


Figure 3. Correlation between total fate factors (i.e sum of grid specific partial Fate Factor (FF) of each region, in ppm.h.yr/kg) and Characterization Factor (CF) for NMVOC and NO_x. Results of the pearson correlation are shown in the top left of the graph.



547

548 **Figure 4. Marginal Effect Factors (MEF), Linear Effect Factors (LEF) and Average**
 549 **Effect Factors (AEF) (with area excluded) for (a) grasslands and (b) forests (left vertical**
 550 **axis). And AOT40 values per grid (right vertical axis) for grasslands and forest plotted**
 551 **as a frequency distribution with steps of 0.02 ppm.h.**